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R & D STATUS REPORT

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SHORT TITLE OF WORK: High Performance YBCO Films

REPORTING PERIOD: Period 5/1/93 to 7/31/93

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SUMMARY

- A number of attempts were made to attach a metal handle consisting of Cu/Stainless Steel #409/Cu composite to YBCO using a crystallizing lead-zinc-borate glass as bonding agent. A strong, well adhering bond was achieved to MgO and $SrTiO_3$ as buffer layers on top of the YBCO. Unfortunately a substantial portion of the buffer layers and the YBCO appear to have been dissolved by the bonding glass. Experiments with thicker buffer layers and other buffer materials are underway. Initial results with Y_2O_3 as a buffer layer look very promising. A glass/metal handle attached to a YBCO/Y_2O_3 layer combination at 460°C showed good adherence and no apparent damage to the YBCO layer. This system is being evaluated further.
- Polishing experiments were conducted on 2.5 x 2.5 cm^2 MgF_2 wafers to establish a process that provides a true epitaxial finish while keeping thickness variations to within $\pm$ 1μ.
- High resolution transmission electron microscopy (HRTEM) and selective area electron diffraction studies (SAED) were performed on YBCO/buffer layer/substrate interfaces to study the local crystallographic structure of the layers. These tests which produced very promising results will be used as guidance for attempts to further improve the crystalline quality of the YBCO films.
- The dielectric resonator test setup for measuring the surface resistance of HTS films was modified to determine R_s of YBCO on top of a substrate as well as through the substrate. With this technique we will be in a position to judge if there is any deterioration in R_s after the handle attachment and subsequent processing steps.

I. INTRODUCTION

As discussed in more detail in the Last Quarterly Report the main emphasis of this program has been shifted towards multichip module (MCM) requirements. The successful deposition of YBCO on MgF_2 (a low-ε substrate) achieved in the early part of this program opened the way for an *alternative* approach to the superconductive interconnect problem for MCM assemblies. The new approach depends on the ability to apply a metal handle to the YBCO film deposited on MgF_2 and subsequently thin and polish the MgF_2 to a layer 5 to 10μm thick. Subassemblies of this type promise to be very useful for millimeter wave applications as well as MCM interconnects.

The majority of the effort under this program was therefore divided into 3 areas: the further improvement of HTS film quality on MgF_2, the attachment of a metal handle to the top YBCO layer and the thinning of MgF_2 to very tight tolerances. This work was complemented by experiments on necessary buffer layers between the YBCO and the glass/metal handle and development of measurement techniques that permit assessment of the quality of the HTS film before and after handle attachment and subsequent processing.

II. HTS MATERIALS DEVELOPMENT

1. High Resolution Transmission Electron Microscopic and Selective Area Electron Diffraction Studies on YBCO - $SrTiO_3$ - MgO - MgF_2 multilayers

The interface between the high T_c film (YBCO) and the substrate over which the film is deposited is one of the important factors that can strongly influence the microwave and millimeter wave characteristics of a patterned superconducting film. In the present context of YBCO films on MgF_2 substrates with MgO and $SrTiO_3$ buffer layers, study of the *film - buffer layer - substrate* interface is important to refine the film deposition technology to further lower the surface resistance of the YBCO films. In this quarterly report, we summarize our High Resolution Transmission Electron Microscopy (HRTEM) and Selective Area Electron Diffraction results of YBCO-$SrTiO_3$-MgO-MgF_2 multilayers aimed a investigating the film-buffer layer-substrate interfaces.

The HRTEM investigations of the interfaces were carried out in the cross section mode. The specimen were prepared by standard specimen preparation techniques, including mechanical lapping and ion milling with liquid nitrogen cooling. We also carried out Selective Area Electron Diffraction (SAED) studies to investigate local crystallographic structure of the substrate-buffer layer as well as film (YBCO)-buffer layer-substrate interfaces. We employed special apertures for carrying out SAED work.

A typical HRTEM lattice image of the epitaxially grown YBCO film on MgF_2 substrates with $SrTiO_3$ and MgO buffer layers is shown in Fig. 1. The epitaxial nature of the YBCO film and the underlying $SrTiO_3$ buffer layer is inferred from their lattice images in the figure. It is notable that the interface between the YBCO film and the $SrTiO_3$ buffer layer is rather sharp with no detectable secondary phases at the interface. The small amount of waviness seen in the YBCO layers could be due to the presence of crystallographic stacking faults. A further refinement in the deposition process is presently being carried out to improve the crystalline quality of the YBCO films.

In Fig. 2 we present cross sectional HRTEM bright field image of a typical YBCO-$SrTiO_3$-MgO-MgF_2 multilayer along with the SAED data obtained from the MgF_2 substrate (bottom left), MgF_2-MgO interface (top left) and MgF_2-MgO-$SrTiO_3$-YBCO interface (top right). All the diffraction patterns were registered along the [010] zone axis. The results of these diffraction studies can be summarized as follows.

(001) MgF_2	∥ (001) MgO;	(100) MgF_2	∥ (100) MgO;
(001) MgO	∥ (001) $SrTiO_3$;	(100) MgO	∥ (100) $SrTiO_3$
(001) $SrTiO_3$	∥ (001) YBCO;	(100) $SrTiO_3$	∥ (100) YBCO

The orientation crystallographic relationships between the various layers and the substrate indicate a **cube on cube** epitaxy of all the individuals layers, indicating an achievement of high quality superconducting YBCO films.

The data obtained in these studies is complimentary to the x-ray structural information previously presented in one of our earlier reports (October '92). In the x-ray diffraction studies however, it is not possible to obtain the interfacial information. The present HRTEM and SAED investigations with the unique ability to probe local microscopic structural information are instrumental in our refining the deposition technology of the buffer layers as well as YBCO on the MgF_2 substrates.

Fig. 1. Cross sectional High Resolution Transmission Electron Microscope lattice images of $YBCO/SrTiO_3/MgO/MgF_2$ multilayers.

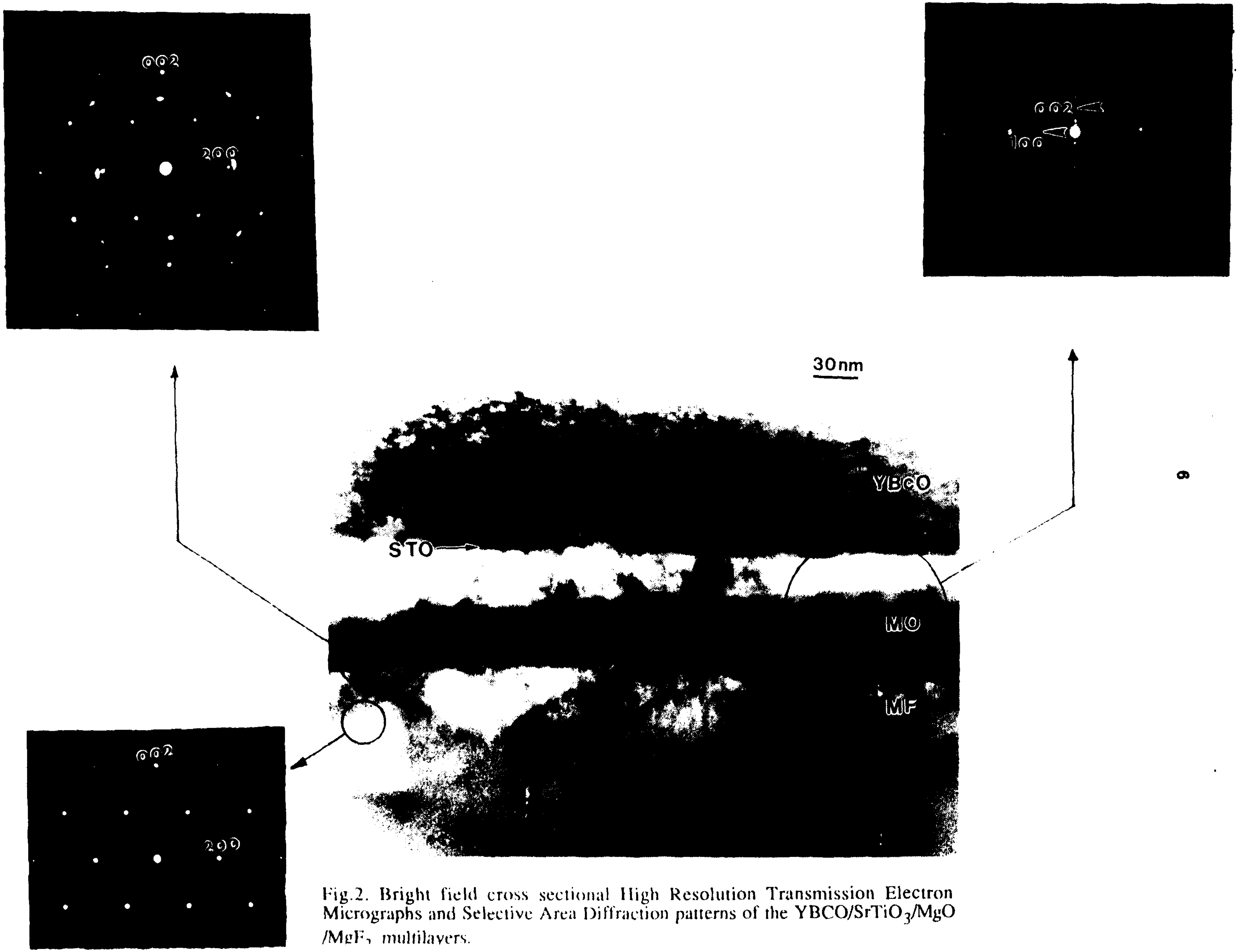

Fig.2. Bright field cross sectional High Resolution Transmission Electron Micrographs and Selective Area Diffraction patterns of the YBCO/$SrTiO_3$/MgO /MgF_2 multilayers.

2. Buffer layers for glass/metal handle attachment

a) Yttrium Oxide

Currently, the glass handle experiments are being carried out on $SrTiO_3$-MgO-YBCO-$SrTiO_3$-MgO-MgF_2 multilayers. For obtaining the best results, the top surface ($SrTiO_3$ in this case) to which the glass is attached should act as an ideal buffer layer, preventing any possible chemical reaction of glass with YBCO by diffusion. Even though $SrTiO_3$ could be effective as a chemical barrier for any possible reaction of glass with YBCO, recent experiments carried out by us point out that Yttrium Oxide (Y_2O_3) could be even a better choice. Y_2O_3 is chemically and structurally compatible with YBCO and we have epitaxially grown Y_2O_3 over YBCO at the same growth conditions, without deteriorating the superconducting properties of the YBCO film. In Fig. 3 (a) we present the AC susceptibility data on one typical film showing a T_c of ~ 89K. During our investigations we also found that the adhesion of Y_2O_3 to glass is excellent even at relatively low temperatures (200°C). These results suggest the possibility of using Y_2O_3 as a cap layer for the on-going glass handle experiments. These possibilities are currently being evaluated.

b) Silver

Recently, Pond et al. [1] reported using a Silver (Ag) film as cap layer over YBCO in obtaining two sided YBCO deposition. Their experiments suggest a possibility of using Ag as a cap layer for the glass handle experiments as well. We have deposited Ag over the YBCO - $SrTiO_3$ - MgO - MgF_2 multilayers and the AC susceptibility data of one typical structure is presented in Fig. 3 (b). The YBCO in this multilayer structure exhibits a T_c of ~ 89 K. The deposition technology is being refined. These approaches using different cap layers YBCO are carried out in an attempt to evaluate the best scheme for the glass handle experiments.

III. SUBSTRATE/HANDLE DEVELOPMENT

1. Glass/Metal Handle Attachment

The glass/metal handle for attachment to YBCO films currently under development at Sarnoff is shown in Fig. 4. The handle will be composed of a thick (30 mils) metal base (Cu/Stainless Steel #409/Cu) with a relatively thin (0.5 - 1 mil) bonding layer glass (a crystallizing lead-zinc-borate glass is used) applied to it by

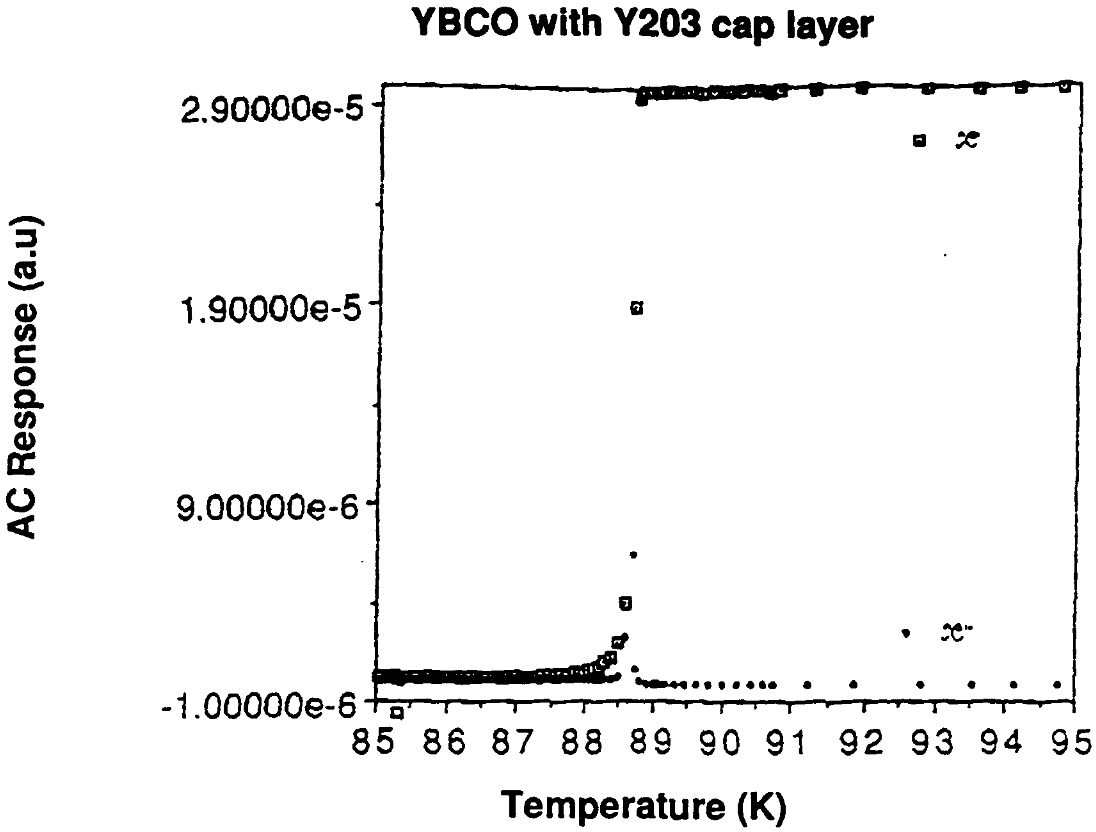

Fig. 3(a). AC Susceptibility data of a typical epitaxial Y_2O_3/YBCO/$SrTiO_3$/MgO/MgF_2 multilayer.

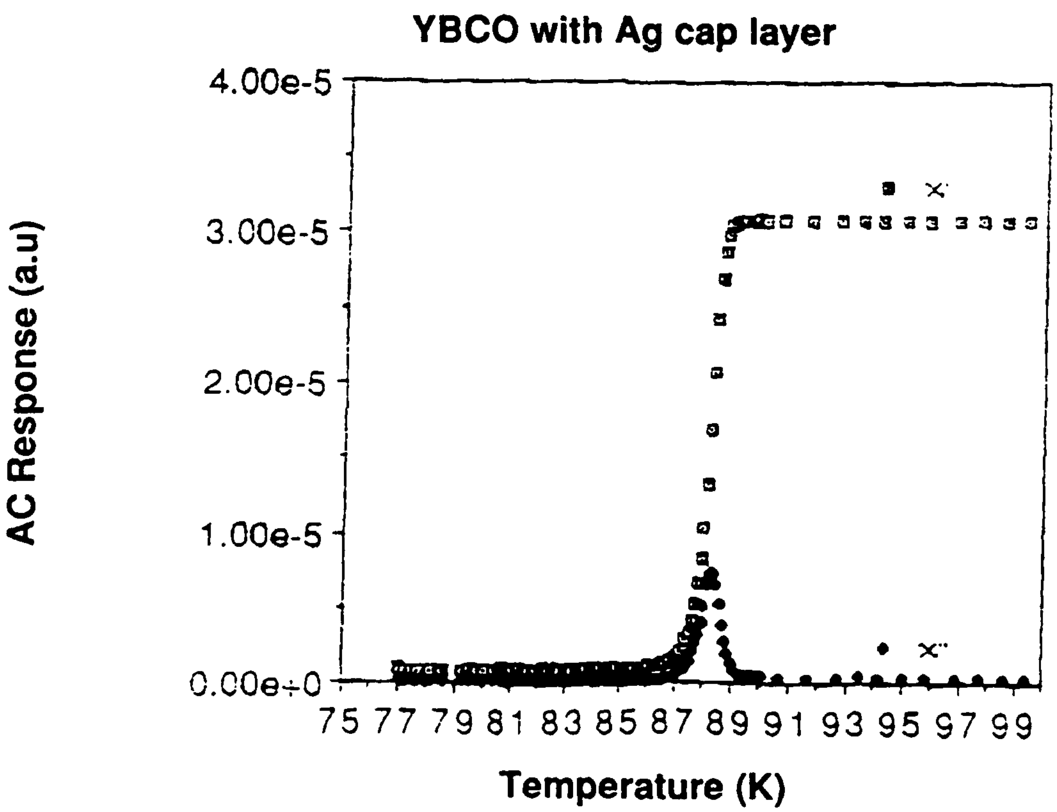

Fig. 3(b). AC Susceptibility data of a typical Ag/YBCO/$SrTiO_3$/MgO/MgF_2 multilayer thick film screening techniques. The bonding layer is then fired onto the metal base at a 460°C peak temperature in air. Attachment of the YBCO film to the glass/metal handle follows with a separate firing between 475 and 600°C in air. A buffer layer is required, otherwise the YBCO film will dissolve into the bonding glass during handle attachment.

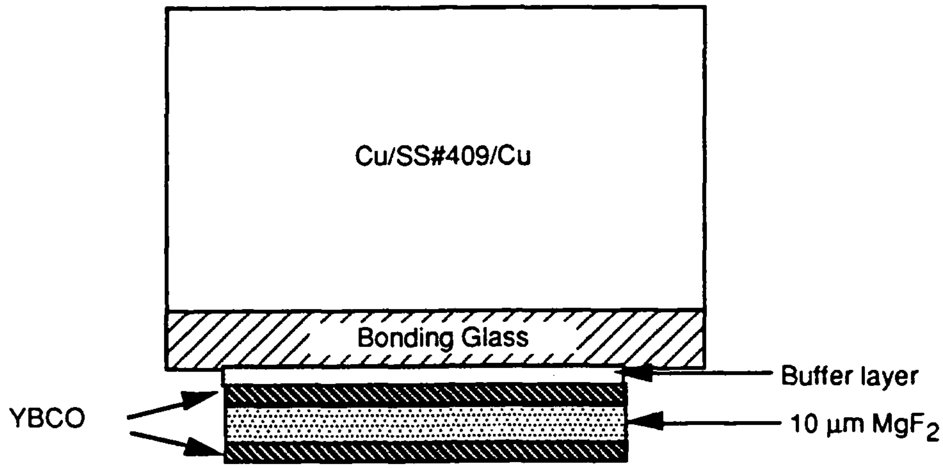

Fig. 4: Glass/metal handle structure (not to scale)

The first bonding experiments were performed on YBCO samples, each having a thin (2000Å) buffer layer composed of MgO covered with $SrTiO_3$, that were supplied by Neocera. During the glass/metal handle attachment process, the buffer layer (and some of the YBCO film) appeared to have been dissolved by the bonding glass. However,

- the bond to the YBCO film was strong;
- the sample did not exhibit any signs of cracking, indicating that the MgF_2, bonding glass, and Cu/SS#409/Cu are reasonably well thermal-expansion matched.

A second buffer layer material, a 2000Å Ag film, was also tested. This also did not prevent the YBCO from being dissolved by the bonding glass layer. To limit the rate of dissolution of the Ag film into the bonding layer glass, several bonding layer formulations containing silver flake (15 - 70 wt.%) were formulated, printed and fired on metal handles. However, the silver flake reduced the flow properties of the bonding layer, resulting in a rough surface after firing onto the metal. A similar result was obtained when fused MgO was added as an inert filler to the lead-zinc-borate glass. The rough surface, caused by these fillers, only allows the formation of point contact bonds when the glass/metal handle is attached to the YBCO film. These point contact bonds are too weak for reliable handle attachment. Use of fillers in the bonding layer will require polishing, prior to handle attachment, to obtain an adequate surface for a high strength joint.

The best results to date have been achieved with a Y_2O_3 buffer layer, approximately 7000Å thick. This material was in situ deposited on YBCO in the vacuum chamber at Neocera. A Cu/Stu Su/Cu handle attached to the buffer-layer/YBCO combination at 460°C showed good adherence and apparently no damage to the YBCO. The initial tests were performed under 40X magnification. A second series of tests will not measure the YBCO surface resistance (see also section III.3) before and after handle attachment and will also explore any possible deterioration at the higher temperatures required for the subsequent growth of a second YBCO layer.

During the upcoming quarter, our main effort will thus be diverted towards refining high quality, impervious buffer layers on top of the YBCO film. Aside from further exploring Y_2O_3 we will also investigate increasing the thickness of the $MgO/SrTiO_3$ layers to ~1μm. A silver film can also be laser-deposited on top of the $SrTiO_3$, if required. The quality of the buffer layer will be determined by optical microscopy through the substrate layer and by measurement of the superconducting properties of the YBCO after attachment of the glass/metal handle.

To increase the availability of samples for developing a high quality buffer layer, the initial experiments will be performed on YBCO films deposited on $LaAlO_3$ substrates, which are easier to fabricate. After promising results have been obtained, the results will be verified on YBCO films deposited on MgF_2 substrates.

The fabrication process for glass/metal handles will also have to take into account the subsequent polishing of the MgF_2 substrate to thicknesses below 10μm. This requires that the glass/metal handle be fabricated to exacting tolerances; both surfaces of the 30 mil (760μm) thick handle must be parallel to within ±1μm. Such tolerances cannot be routinely obtained from commodity products, without additional processing. It is expected that the additional fabrication steps depicted in Fig. 5 will be required to make the Cu/SS#409/Cu laminate suitable for use in the glass/metal handle. Cu/SS#409/Cu is produced by

Texas Instruments in several thousand pound rolls. The metal handles must be cut or stamped from these rolls. and then all camber must be removed by a suitable heat treatment process, that does not heavily oxidize the Cu surface. After flattening, the Cu surface must then be roughened (by etching), so that the lead-zinc-borate glass will strongly bond to it. Following application of the bonding glass layer, the backside of the metal handle must be polished flat and parallel to the top side. This will be required so that the MgF_2 substrate can be thinned to 10μm.

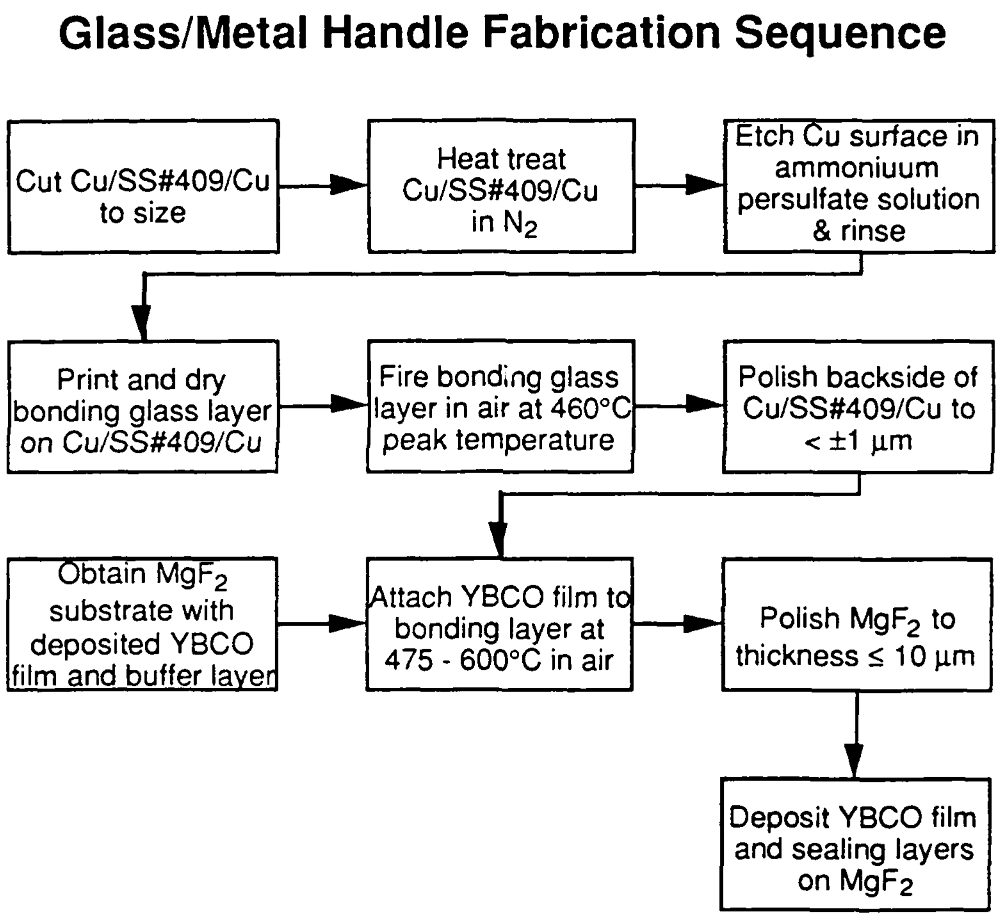

Fig. 5. A process diagram for glass/metal handle fabrication and attachment to the YBCO film/MgF_2 substrate

2. MgF_2 Thinning and Polishing

Last quarter, we experimented with reducing the thickness of small MgF_2 samples, 0.635 x 0.635 cm^2. Due to their small size, those samples were lapped first on a tin lap embedded with diamond to remove as much material as possible.

This quarter our efforts concentrated on improving the manufacturer's polish of complete 2.54 x 2.54 cm square single crystal MgF_2 wafers. Past experience with YBCO deposition on MgF_2 has shown the importance of starting with a very high quality epitaxial finish on the MgF_2 wafers and the finish supplied by various manufacturers is less than ideal. As a result of the larger size of this quarter's samples we could start with a mechanical polish and use 1 µm alumina polish. This first step is necessary to eliminate any differences in thickness between the samples. (Samples are polished in groups of three.) The larger pieces are then chemically polished with Rodel semiconductor polishing slurry. This resulted in a scratch free and sleek free surface. Sleeks are micro cracks which are only visible under high magnification. A sleek free surface finish is classified as epitaxial. This method is also applicable for larger size samples. The test wafers with improved polish will be evaluated by Neocera by depositing YBCO films on thin.

The final procedure for preparing substrates for glass handle bonding is detailed below. The samples, as received from the manufacturer, are first inspected for scratches and micro cracks. Using the above mentioned process, the samples are then polished to an epitaxial or sleek free finish. The thickness of the samples is not measured at this point to avoid marring the surface. The YBCO and appropriate buffer layers are then laser deposited. At this point we will measure the thickness because the buffer layer protects the YBCO on the top surface and the bottom surface will be repolished after the handle is applied. Now the MgF_2 can be lapped and polished to the desired thickness of 10µm. By knowing the total thickness of the MgF_2 sample we do not need to know the exact thickness of the glass.

3. Measurement of R_s in the presence of a metal handle

In developing the glass/metal handle for attachment to YBCO, there is a concern about the status of the superconducting material at the end of this process. The effect of the bonding layer, processing temperature etc., can degrade the rf performance of the previously deposited layer. AC susceptibility testing is normally used for checking the HTS film after deposition, however, the presence of this glass/metal handle makes it difficult to obtain accurate results, especially if the metal (in our case Cu/SS#409/Cu) is slightly magnetic.

Our dielectric resonator (DR) test setup is suitable to test the superconducting material before and after attaching the handle. The DR setup can measure the top and bottom surfaces of a superconducting layer deposited on one side of a substrate. The DR can measure the top surface when it is used as an end plate of the DR cavity (top side UP), which is the normal mode of operation. The DR also can be used to evaluate the bottom surface if it is placed (upside-DOWN) as an end plate. Of course, including the substrate material (MgF_2) inside the cavity as a bottom layer between the DR puck and the superconducting ground plane will affect the resonant frequency, and the measured Q due to the change of cavity dimensions, filling factor, and the effect of metallization on the cavity's Q.

We have carried out a sample experiment to measure the surface resistance of a superconducting layer deposited on one side of $LaAlO_3$. We used it as end plate of the DR resonator and measured the center frequency and the Q for both cases (top side up, and topside down). The measured Q and center frequency in both cases were used to evaluate the surface resistance of the sample. As was expected, the center frequency of the top side down case is lower and the Q is higher. Close agreement between the two calculated values (of R_s) was seen. Fig. 6 shows the resonant frequency and the Q as function of temperature. The calculated surface resistance value is 1m Ω at 77K. This technique thus can be used to evaluate HTS film properties before and after handle attachment process steps have been performed.

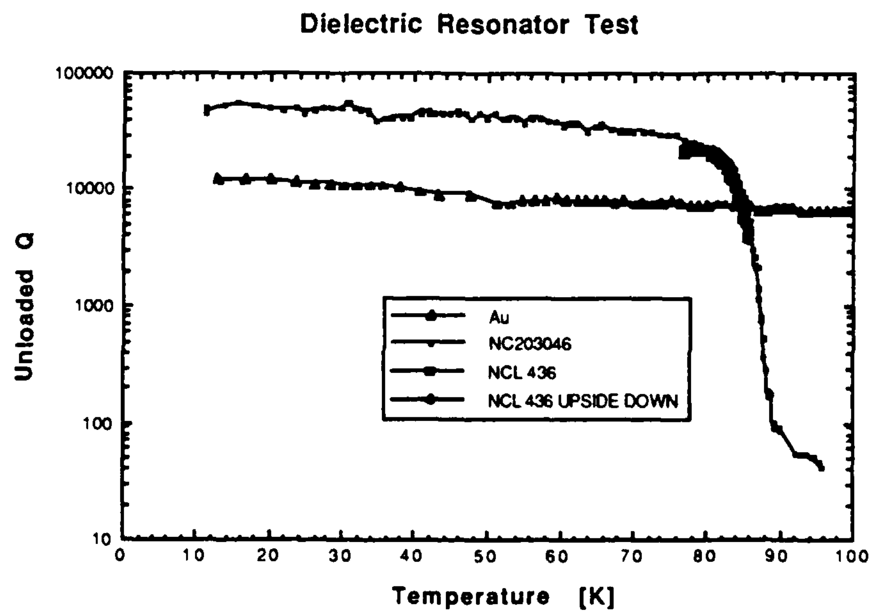

Fig. 6.

IV. REFERENCE

1. J. M. Pond et al., IEEE Trans. on Applied Superconductivity, 3, 1711 (1993).

V. CHANGE IN KEY PERSONNEL: None

VI. SUMMARY OF SUBSTANTIVE INFORMATION DERIVED FROM SPECIAL EVENTS: None

VII. PROBLEMS ENCOUNTERED AND/OR ANTICIPATED:

The buffer layers on top of YBCO investigated so far: $MgO/SrTiO_3$ and Ag were not sufficient to prevent glass YBCO interaction. At present it is not clear if this interaction is fundamental or caused by defects in the buffer layer that are penetrated by the glass under high temperature processing. The processes that provide the initial promising results using layers such as Y_2O_3 and thicker $SrTiO_3$ are being investigated further.

VIII. ACTION REQUIRED BY THE GOVERNMENT : None

IX. FISCAL STATUS

1. Amount currently provided on contract:	$900K
2. Expenditures and commitments to date:	$727K
3. Funds required to complete work:	$1,585,085